Galer, Rose

From:	Rose, Jay
Sent:	Tuesday, December 18, 2007 9:51 AM
To:	Galer, Rose
Subject:	FW: todosow 2007
Importance:	High
Attachments:	Chapter 3 Info-REVMT.doc

From: Michael Todosow [mailto:todosowm@bnl.gov]
Sent: Wednesday, October 17, 2007 11:28 PM
To: Rose, Jay; francis.schwartz@hq.doe.gov
Cc: Roald Wigeland; Phillip J Finck
Subject: PEIS thorium
Importance: High

Here's a stab at updating sections in Chapter 3 to be consistent with the concepts described in Section 2. I'm not comfortable with the part on Pu-burning...will need to check details tomorrow. We should settle on a way to refer to the required uranium enrichment: what will be required are enrichments "up to the nonproliferation of 20 w/o". I gave up on changing all the places where this occurs, and have highlighted them **"in yellow"**.

My gut feeling is that the Appendix either repeats things that have been said before, or is way more information than anybody wants or needs to know in the context of the PEIS - I would vote to drop it (maybe I'll see some redeeming features with fresh eyes tomorrow).

```
*********************************************************
* Michael Todosow          * Phone: (631) 344-2445   *
* Brookhaven National Laboratory  * FAX: (631) 344-7650    *
* Building 475B            * E-Mail: todosowm@bnl.gov *
* 12 S. Upton Rd.          *                *
* P.O. Box 5000            *                *
* Upton, NY 11973-5000          *              *
*********************************************************
```

12/20/2007

3.3   THORIUM ONCE-THROUGH FUEL CYCLE ALTERNATIVE (THORIUM ALTERNATIVE)

The Thorium Alternative, described in Section 2.2.3.3, would represent a fundamental shift in the fuel used for U.S. commercial reactors. Rather than being fueled solely by enriched (3-5%) uranium, U.S. commercial reactors would transition to a fuel composed of thorium and enriched uranium (<20%), but would continue to operate using a once-through fuel cycle to produce electricity. Of the possible implementation options, preliminary studies suggest that the most attractive approach is a heterogeneous, seed-blanket configuration along the lines of the seed-blanket-unit (SBU), or whole-assembly-seed-blanket (WASB) concepts described. These concepts involve two distinct zones in a reactor: a seed region containing uranium oxide fuel with the uranium enriched up to ~19.9 w/o, and a blanket region containing thorium oxide and approximately 10 v/o uranium oxide, where the uranium enrichment can range up to ~19.9 w/o. These concepts therefore include characteristics of both conventional uranium fuels, albeit with a significantly higher enrichment than in current commercial reactors, and thorium based fuels. At the programmatic level, this PEIS assesses the potential environmental impacts associated with broad implementation of the Thorium Alternative. The analysis of this broad implementation assumes that the U.S. commercial reactors begin to shift to thorium-based fuel concepts by approximately 2022, and that by 2030, all commercial reactors would operate using thorium-based fuels. Additionally, in order to provide a comparable basis among the alternatives, the PEIS assesses the environmental impacts of producing 100 GWe using a thorium-based once-through fuel cycle. That comparative analysis is presented in Section 3.6.

| | Deleted: s |

This PEIS presents the environmental impacts of the Thorium Alternative as follows:

- **SNF and HLW generated up to the Yucca Mountain statutory limit:** these impacts would be the same as those presented in Section 3.1.1 and are not repeated.
- **Thorium-Based Facility Operations:** the thorium fuel cycle would not necessarily require new facilities (although this PEIS includes the option of constructing a new fuel fabrication facility). Rather, existing facilities would operate differently using a thorium fuel cycle. At the front-end of the fuel cycle, thorium would need to be mined and there would be a minor reduction in natural uranium requirements. These impacts are presented. With respect to uranium enrichment and fuel fabrication, a thorium fuel cycle would also operate differently than the uranium fuel cycle. The impacts of producing higher enriched uranium fuel (up to ~19.9%) are presented. Reactor operations using thorium-based fuel are also discussed, including the impacts associated with establishing a hypothetical geologic repository for SNF from the thorium fuel cycle and the transport and emplacement of these wastes in that hypothetical repository. These impacts are presented in Section 3.3.1.
- **New nuclear electricity generation in 2010-2060:** the environmental impacts of producing up to 111-239 GWe of additional electricity in nuclear reactors, including the construction and operation of 111-239 new reactors, would be the same as those presented in Section 3.2.2 and are not repeated.
- **Displacement of non-nuclear electricity generation:** for the reference case regarding electricity growth, the environmental impacts of displacing approximately 100 GWe of non-nuclear electricity capacity with nuclear power would be the same as those presented in Section 3.2.3 and are not repeated.
- **Ongoing nuclear fuel cycle R&D activities, including AFCI:** these impacts would

be the same as are presented in Section 3.1.5 and are not repeated.

3.3.1   Thorium-Based Facility Operations

This section discusses operation of the thorium-based once-through fuel cycle. Mining, enrichment, fuel fabrication, and reactor operations are all addressed.

Thorium Mining. Thorium is relatively abundant and easily mined. Monazite, a mixed thorium rare earth uranium phosphate, is the most popular source of thorium and is available in many countries (particularly India, Australia, and Brazil) in beach or river sands along with heavy minerals– ilmenite, rutile, monazite, zircon, sillimenite and garnet. The present production of thorium is almost entirely as a by–product of rare earth extraction from monazite sand. The mining and extraction of thorium from monazite is relatively easy and significantly different from that of obtaining uranium from its ores. For example, the overburden (the soil and rock above the deposit) during mining is much smaller than in the case of uranium and the total radioactive waste production in mining operation is about 2 orders of magnitude lower than that of uranium. The potential radiological impact to miners is also much smaller than in the uranium case due to the short lifetime of thoron (predominant radon in the thorium, Rn-220 with a half-life of 56-sec.) as compared with the predominant radon in the uranium ore (Rn-222 with a half-life of 3.8 days), and needs therefore, much simpler tailings management than in the case of uranium, to prevent long term public doses (see Figure 3.3.1-1) (ref.). External gamma exposure is not a major concern because thorium emits only a small amount of gamma radiation. Consequently, thorium is generally a health hazard only if it is taken into the body. If inhaled, thorium can have approximately 8 times a greater health risk than natural uranium. The main health concern for environmental exposures is generally bone cancer.
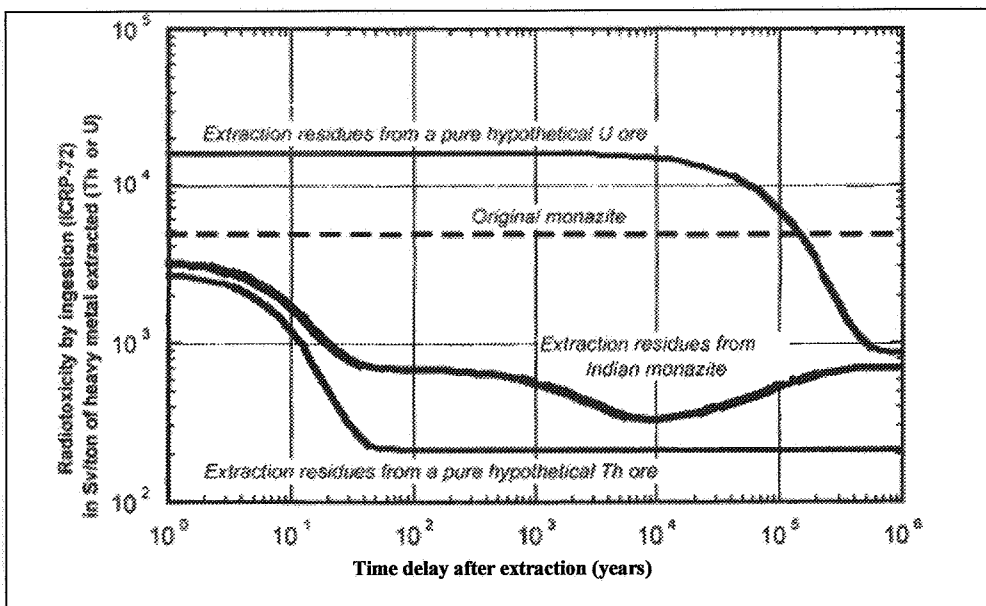
FIGURE 3.3.1-1—*Radiotoxicity of Uranium Ore versus Thorium Ore*

Thorium Fuel Enrichment and Fuel Fabrication.   The environmental impacts of enriching uranium to 19.9% and fabricating fuel for thorium-fueled reactors would be similar to the impacts described in Section 3.1.3.1.2. More details regarding uranium enrichment

and thorium fuel fabrication are contained in Appendix A. The thorium fuel cycle would require uranium enrichments of up to ~19.9% versus the 3-5% for the uranium fuel cycle. Based on the SBU concept described in Section 2.2.3.2, a typical thorium-fueled LWR (1 GWe) would require:

- Core loading would contain 42 tons of Th and 7 tons of uranium (in the 193 blanket subassemblies) and 20 tons of enriched uranium (19.9%) in the seed subassemblies;
- To produce 20 tons of 19.9% enriched uranium, the facility would need about 800 tons of natural uranium;
- Natural uranium requirements normalized to GWe-year would be 199 tons for the thorium-fueled reactor (versus 203 tons for the uranium-fueled reactor) (Ref.)

Currently, there is no capacity in the U. S. to enrich uranium to 19.9%. While the technology exists and has been utilized in the past to produce uranium with enrichments of 19.9% (and higher), an existing enrichment facility would need to be retrofitted (with additional centrifuges connected in series or additional gaseous diffusion stages) or a new facility constructed. Such a new facility would likely be a large industrial facility similar in nature to the facilities that were used in the past (i.e. K-25, Portsmouth, Paducah). In general, these facilities required hundreds of acres, used significant quantities of electricity, and employed thousands. Modern enrichment facilities would likely be more compact, and more efficient in terms of electricity and staffing. The size of an enrichment facility is generally a compromise between criticality concerns (which govern the size of components), and desired enrichment and throughput. For example, multiple passes through enrichment stages can be used to increase the enrichment, subject to criticality constraints. In addition, the RERTR (Reduced Enrichment for Research and Test Reactors) program also needs uranium enriched up to 20 w/o to replace the HEU fuel currently employed in many of these reactors. The option of obtaining these enrichments by down-blending surplus HEU from the weapons complex may be available to satisfy some of the requirement. Section 3.1.3.1.2 discusses the types of impacts associated with these facilities.

Thorium fuels have been made in the past, and for oxide, one presumably suitable technology would be similar to that already established industrially for uranium oxide and MOX fuels formed from pellets in tubular cladding. A separate plant may be needed to avoid cross contamination (or dedicated lines in existing facilities may be adequate), and the optimum conditions could well be rather different, but no serious difficulties seem likely for once-through applications. If interest passes to nitride or other less familiar forms, then industrialization is likely to need a more radical development program. No special problems are expected in manufacturing technology of mixed oxide thorium uranium (or plutonium) pelletelized fuel.

Fuels containing naturally occurring 'fissile' U-235 in combination with 'fertile' U-238 or Th-232, emitting only alpha particles of relatively low specific activity, can be manufactured by the so-called 'contact operations' where the operator has direct contact with the fuel material. However, process operations that involve generation and handling of fine powders of U-235, U-238, or Th-232 bearing fuels are carried out in ventilated enclosures, such as glove boxes, for minimizing radioactive aerosol.

As discussed in Section 3.1.3.1.3, there are only two fuel fabrication facilities in the U.S. that are licensed to fabricate reactor fuels with enrichments greater than 5%. These two facilities produce fuels for the naval reactors program mainly, as well as research reactor fuels. Because the capacity of these two fuel fabrication facilities would not be sufficient to produce all of the up to 19.9% enriched uranium fuel for the commercial industry, it is likely

that one or more new fuel fabrication facilities would be constructed.

Additionally, the existing facilities are designed to handle and produce highly enriched uranium fuel, which would lead to a number of physical restrictions to avoid criticality. These would not be nearly as restrictive for a facility handling 19.9% enriched uranium. Lastly, thorium-specific hazards (such as greater risks from inhalation) would need to be accommodated in the design of the fuel fabrication facility. These would likely not be present in a facility designed for highly enriched uranium fuels, and backfitting would likely be undesirable from cost and other considerations.

Reactor Operations. Although there would be changes in reactor operations, it is expected that the design would ensure that all safety and operational parameters of existing nuclear plants would be preserved with the seed-blanket options(Ref). For the seed-blanket arrangement, although the maximum local power density in the seed part of the assembly could be about 1.3 times higher than that of the typical PWR fuel, no critical heat flux margins would be exceeded (Ref). Reactor-specific designs and operating procedures could also be employed to ensure margins are maintained. For example, the seed material could be replaced more often and/or reshuffled more frequently, similar to conventionally fuelled uranium reactors. Issues associated with the increased reactivity due to continuing production of U-233 from the decay of Pa-233 following shutdown need to be taken into consideration but should not be a practical concern. However, the additional U-233 that will be produced as Pa-233 decays needs to be accounted for in satisfying the non-proliferation limit discussed in Section 2. In theory, longer refueling cycles and higher plant capacity factors could be achieved with thorium fuel because thorium fuel has a significantly higher thermal conductivity at LWR operating temperatures and a lower rate of fission gas release. Therefore, thorium fuel can be operated to higher burnup with less difficulty than uranium fuel. However, as noted earlier, the uranium-based seed fuel may limit the potential gains in cycle length.

In general, the following plant parameters could change for a concept employing thorium-based fuel; statements related to advantages due to the properties of thorium oxide, however, are only applicable to the blanket portion of seed-blanket configurations;

Land Use: overall land use would not change appreciably, with the possible exception of expanded pool storage, which might be required to accommodate the longer cooling times of thorium fuels due to higher initial decay heat. Because spent fuel storage pools are a relatively small portion of the total land area associated with reactors plants, this impact is not expected to be major.

Water Use: cooling water requirements are largely a function of reactor power and thus, would not be affected by the thorium fuel cycle.

Effluents and Emissions: Thorium-based fuels are expected to have superior thermo-physical properties, such as higher melting point, better thermal conductivity, and lesser release of fission gas as compared to uranium-based fuels. As such, for the same burnup, effluents and emissions should re reduced.

Operating Personnel: No changes expected.

Worker Health: Irradiated thorium-based fuels contain a significant amount of U-232,

---

Deleted: thorium fuel cycle

Deleted: Additionally, in the conversion chain of Th-232 to U-233, protactinium-233 (Pa-233) is formed, which could pose a problem on reactor shutdown.[1] By contrast, the Np-239 intermediate between the U-238 and Pu-239, has a half-life of 2.3 days.

Deleted: cycle

Deleted: [2]

which has a half-life of only 73.6 years and is associated with strong gamma emitting daughter products. As a result, there is significant buildup of radiation dose with the storage of spent thorium–based fuels. As a result, operational doses could be higher for storage workers for the thorium fuel cycle. For the uranium fuel cycle, the MEI dose for an involved storage worker would be approximately 170 mrem/yr (see Table 3.2-4). Even if this dose were to double, the resultant LCF risk would be less than 0.20. Paradoxically, this disadvantage could be considered as an advantage from the non-proliferation point of view.

Spent Nuclear Fuel. The thorium fuel cycle offers several potential advantages relative to the conventional uranium fuel cycle, including: (1) reducing the quantity and quality of plutonium produced; (2) producing fewer minor actinides (Np, Am, and Cm), which minimizes long-term toxicity and decay heat; and (3) improving the long-term SNF waste characteristics. These advantages are further explained below.

Plutonium Produced. Table 3.3.1-1 presents the characteristics of the plutonium produced by a thorium fuel versus a typical PWR. As can be seen from that table:

- Total plutonium production is a factor of 3-4 less in thorium fuel than in uranium fuel due to the higher enrichment in the seed and the thorium in the blanket.
- Pu-239 production is a factor of 4.2 less in thorium fuel than in uranium fuel.
- The plutonium produced in the thorium fuel and in the seed is high in Pu-238, leading to a decay heat rate 3.7 times greater than that from plutonium derived from uranium fuel and 29 times greater than that from weapons grade plutonium. The decay heat in thorium SNF is high enough to complicate potential use in a weapon.

TABLE 3.3.1-1—*Plutonium Produced in Uranium-Fueled PWR versus Thorium-Fueled*

Plutonium Isotope	Typical Present-Day PWR		Typical SBU Seed + Blanket		Typical WASB Seed + Blanket	
	kg/GWe-yr	Fraction of Pu (%)	kg/GWe-yr	Fraction of Pu (%)	kg/GWe-yr	Fraction of Pu (%)
238	7.2	3	5	7	6	8
239	148.8	55	38	47	42	49
240	56.4	21	17	20.5	15	17
241	40.8	15	12	15.5	14	17
242	16.8	6	8	10	8	9
Total	270	100	80	100	85	100

For thorium fuels, a large reduction in the discharged fuel mass (about 70 percent) and volume (about 50 percent) could be realized (ref).

Producing Fewer Minor Actinides. Being a lighter element than uranium, thorium fuels produce fewer minor actinides. As a result, the potential radiotoxicity and decay heat of irradiated thorium fuel is generally lower than irradiated uranium fuel. As shown on Figure 3.3.1-2, the level of radiotoxicity is initially much lower than that of uranium fuel. It decays rapidly for the first 1000 years where the radiotoxicity is dominated by Pu-238 and U-232. Beyond this, the dominant isotopes are U-233, Am-241, and Th-229. At 50,000 years the dominant isotopes are Th-229 and Ra-225 and the radiotoxicity is higher than that of conventional uranium fuel.
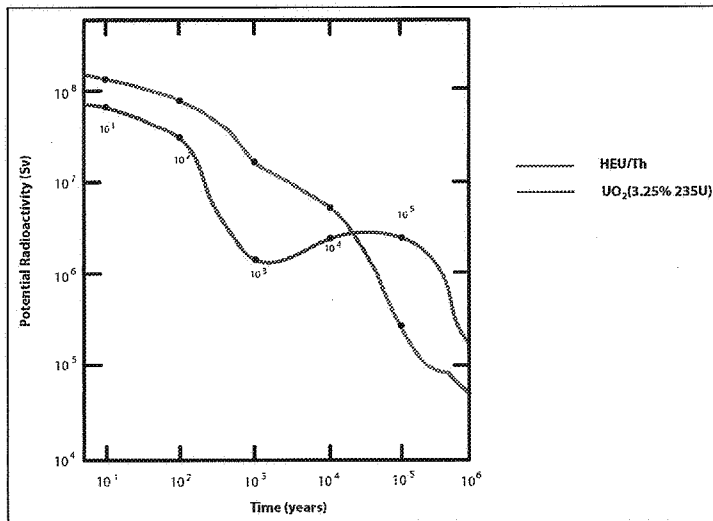
Source: TBD

FIGURE 3.3.1-2—*Radiotoxicity of thorium SNF versus uranium SNF*

Improving Long-term SNF Waste Characteristics. ThO2 is the highest oxide of thorium and does not depart significantly from its stoichiometric composition when exposed to air or water at temperatures up to 2000 degrees Kelvin. Thus, the stability of the oxide form of thorium may help retard the migration of actinides in the repository in case of failure of the clad and other engineered barriers(ref). By contrast, in case of exposure to water, uranium-based SNF fragments react and disintegrate relatively rapidly (about 1 percent per year).

3.3.2 Implementation of the Thorium Alternative

Widespread implementation of the thorium alternative would result in the following domestic impacts:

- Thorium-specific mining (as opposed to by-product mining) would be required;
- Natural uranium needs would slightly decrease compared to the uranium-based fuel cycle (approximately 4 tons less natural uranium per GWe-year);
- Facilities capable of enriching uranium to 19.9% would be required, which could necessitate construction and operation of one or more dedicated enrichment facilities;
- One or more dedicated thorium-uranium fuel fabrication facilities would/could be required;
- Some reactor-related facilities and operations may need to be modified to use thorium-based fuel concepts, which could necessitate construction and operation of additional SNF pools and more robust SNF dry storage facilities;
- Operation of one or more hypothetical geologic repositories would be required for ultimate disposition of thorium SNF. This would also be required for the once-through uranium cycle to accommodate the postulated growth in nuclear power.

Each of these impacts is discussed below. Again, must recognize that the seed-blanket concepts are a mixture of uranium and thorium-based fuels and therefore exhibit the characteristics of both.

Thorium-specific mining. As discussed in Section 3.3.1, in general, the impacts of thorium mining would be less than uranium mining. There would be less overburden, less radioactive

waste produced, less radiological impact to miners, and simpler tailings management than in the case of uranium. However, because the uranium requirements would not be significantly reduced, the thorium-specific mining impacts would be additive. It is noted that the ultimate mining impacts would be highly site-dependent and without any specific mining proposals, quantitative conclusions cannot be made.

Natural Uranium Needs. When normalized to GWe-year, the difference in natural uranium requirements for a thorium-fueled reactor and a uranium-fueled reactor would be approximately 1% (203 tons versus 199 tons). Assuming that nuclear electricity generating capacity would grow to a range of approximately 215–343 GWe by approximately 2060 (see Section 2.2.3.3), the natural uranium requirements would be reduced by approximately 860-1,372 tons annually. This would represent a decrease in natural uranium requirements of approximately 1%. Consequently, the differences are minimal.

Enrichment and Fuel Fabrication Facilities for 19.9% Uranium. Although the specific impacts of constructing and operating one or more enrichment facilities and fuel fabrication facilities specifically to support the thorium cycle are beyond the scope of this programmatic analysis, the following generalities can be made:

The enrichment facilities would be large industrial facilities, similar in size to those discussed in Section 3.1.3.1, with the same types of environmental impacts. In general, enriching uranium to higher than 5% does not produce different types of impacts, but requires more steps.
In order to support a nuclear electricity generating capacity of approximately 215–343 GWe, the Th-U fuel fabrication facilities would need to provide the following capacity throughputs:

- 860-1,375 MT of thorium (based on 42 tons of thorium loading in the blanket and a 10-year in-core residence);
- 4,300-6,860 MT of enriched uranium (based on 20 tons of 19.9% enriched uranium in the seed and a 480-day in-core residence).

Reactor Facilities and Operations. For the thorium fuel cycle, the changes at individual reactor facilities are expected to be minor, as discussed in Section 3.3.1. The most noteworthy change would involve the management of SNF. Because both the discharged volume and mass of SNF would be reduced (by approximately 50 and 70 percent, respectively), there would be less SNF to be managed. For a nuclear electricity generating capacity of approximately 215–343 GWe in 2060, the annual discharged SNF mass would be reduced from approximately 4,300-6,860 (assuming a nominal 20 MTHM SNF per GWe-year) to 1,300- 2,100 MTHM/year. Based on the assumption that all commercial reactors would transition to a thorium-based fuel cycle by approximately 2030, the amount of SNF generated by commercial LWRs over the period of 2010-2060 would range from approximately 73,000 – 87,000 MTHM.

Operation of One or More Hypothetical Geologic Repositories for Thorium SNF. This analysis shows that the thorium fuel cycle, while reducing the mass and volume of SNF, would require a geologic repository capacity on a scale of the Yucca Mountain repository by approximately 2060. The impacts of constructing and operating such a repository would be similar to those of a repository for SNF from the once-through uranium cycle presented in Section 3.1.2.2, and are not repeated here.

3.3.3 Option to Use the Thorium Once-Through Fuel Cycle as a Pu/TRU Burner

In general, the thorium fuel cycle could use mixed thorium plutonium oxide or thorium TRU oxide as a driver fuel. The mixture would be approximately 5% PuO_2 or $TRUO_2$. The exclusion of uranium from the fuel composition would result in an increase in the rate of plutonium incineration compared to the use of standard MOX or TRU fuel (Ref). Studies indicate that up to 1000 kg per GWe-year of reactor grade plutonium could potentially be burned in thorium-based driver fuel assemblies (Ref). Not only would the SNF be degraded in terms of plutonium content, but also would become 'proliferation-resistant' due to the formation of U-232 (from the neutron reaction with Th-232), which has very strong gamma emitting daughter products. In general, the results indicate that the thorium fuel cycle could consume approximately twice the plutonium as conventional MOX fuel with acceptable reactor control and safety characteristics (ref).

If the thorium fuel cycle were used as a Pu/TRU burner, uranium requirements could be reduced to zero during such operations. Assuming a nominal core loading of approximately 1.3 MT/reactor, a 100 MT stockpile of TRU/plutonium could be consumed with 25 reactors every four years. Eventually, TRU/Pu stockpiles would be exhausted, requiring operations with enriched uranium.

Appendix A Info Specific to Thorium

Thorium fuels and fuel cycles offer a number of benefits and challenges for the long term use of nuclear power for electrical generating demand. The benefits include proliferation-resistance, longer fuel cycles, higher burnup rates, improved waste form characteristics, reduction of plutonium inventories and in situ use of bred-in fissile material. These factors have led to the renewed interest in thorium-based fuels and fuel cycles in several developed countries. Thorium fuel cycle is an attractive way to produce long-term nuclear energy with low radiotoxicity waste. In addition, the transition to thorium could be done through the incineration of weapons grade plutonium (WG-Pu) or civilian plutonium.

One of the benefits from introducing thorium into the nuclear fuel cycle is the ability to 'breed' more fissile material. This conversion or 'breeding' process also exists in the uranium fuel cycle, where the isotope of ^{238}U captures fast neutron and through successive decays is converted into the plutonium-239 (^{239}Pu) isotope, which is also a fissile material and can be used as a fuel.

Because the ability of thorium-232 (^{232}Th) to absorb neutrons is nearly three times that of ^{238}U there is a potential for getting a higher conversion ratio, making thorium a better 'fertile' material than ^{238}U in thermal reactors. The use of thorium in the fuel cycle does not eliminate the need for ^{235}U, but does offer the ability to produce new fuel in the form of another, not naturally occurring, isotope of uranium namely uranium-233 (^{233}U). This isotope of uranium is also a fissile material and can produce a fission chain reaction. The combination of ^{232}Th and ^{235}U in a fuel is referred to as the $^{232}Th-^{233}U$ fuel cycle. One advantage of this fuel cycle is the range of neutron energies (speeds) that can be used to produce the fissile isotope ^{233}U. While the production of ^{239}Pu in the uranium fuel cycle relies on fast neutrons the $^{232}Th-^{233}U$ fuel cycle produces ^{233}U at a wide range of neutron energies.

A number of challenges confront the introduction of the $^{232}Th-^{233}U$ fuel cycle into the current commercial nuclear power processes. These challenges include fuel fabrication difficulties that arise because of both thermal chemical properties of thorium as compared to uranium. The melting point of ThO_2 (3,350 °C) is much higher compared to that of UO_2 (2,800 °C). Hence, a much higher sintering temperature (>2,000 °C) is required to produce

high density ThO_2 and ThO_2-based mixed oxide fuels. In addition, since ThO_2 and ThO_2-based mixed oxide fuels are relatively inert and, unlike UO_2 and $(U, Pu)O_2$ fuels, do not dissolve easily in concentrated nitric acid modifications to the fuel processing systems are required. Modifications such as the addition of HF in concentrated HNO_3 to digest the thorium oxide compounds also results in corrosion of stainless steel equipment and piping in reprocessing plants.

Another difficulty with the ^{232}Th–^{233}U fuel cycle is that irradiated Th or Th–based fuels contain significant amount of ^{232}U, which has a half-life of only 73.6 years and is associated with strong gamma emitting daughter products, ^{212}Bi and ^{208}Tl with very short half-life. As a result, there is significant buildup of radiation dose with storage of spent Th–based fuel or separated ^{233}U, necessitating remote and automated reprocessing and re-fabrication in heavily shielded hot cells and increase in the cost of fuel cycle activities.

The conversion process from ^{232}Th to ^{233}U also includes the production of protactinium-233 (^{233}Pa). This isotope ^{233}Pa has a relatively long half-life (~27 days) requiring a longer cooling time of at least one year for completing the decay of ^{233}Pa to ^{233}U. It is essential to separate Pa from the spent fuel solution prior to solvent extraction process for separation of ^{233}U and thorium. Another challenge is that the three stream process of separation of uranium, plutonium and thorium from spent $(Th, Pu)O_2$ fuel, though viable, is yet to be developed.

Thorium and Uranium Fuel Cycles

Nuclear materials consist of fissile materials, which produce a net increase in neutrons when they absorb neutrons, and fertile materials, which produce fissile material when they absorb neutrons. The principal fissile materials for commercial nuclear power are ^{235}U, ^{239}Pu, and ^{233}U. The principal fertile materials are ^{238}U and ^{232}Th. The only fissile isotope to occur in nature in significant quantity is ^{235}U at about 0.711 w/o with ^{238}U being approximately 99.283 w/o and ^{234}U as a nearly negligible trace constituent at about 0.0058 w/o. The fertile isotope ^{238}U is converted to the fissile isotope of plutonium-239 (^{239}Pu) after absorbing a neutron and undergoing several subsequent decays. The process by which an initial fuel containing approximately 3 to 5 w/o ^{235}U and 95 to 97 w/o ^{238}U is burned in a reactor and producing 239Pu from the transmutation of ^{238}U for producing a sustained nuclear chain reaction is commonly referred to as the uranium fuel cycle.

The thorium fuel cycle on the other hand is the process by which an initial fuel containing ^{235}U or a mixture of ^{235}U and ^{239}Pu (mixed oxide fuels or MOX) along with the fertile ^{232}Th is burned in a reactor thereby producing a new fissile material ^{233}U, through the transmutation of the thorium into uranium, for producing a a sustained nuclear chain reaction. Thorium-232, which occurs naturally at approximately a 3 – 4 percent metal in ores, is also a fertile material that can be converted to the fissile isotope of ^{233}U by absorption of a neutron and successive decays.

Uranium and Thorium Mining

Thorium Mining:

The major commercial source of thorium is monazite, an anhydrous rare-earth phosphate with the chemical formula $(Ce, La, Nd, Th) PO_4$. Typically, 3 to 5 percent of the metal content of monazite is thorium (in the form of thorium dioxide, ThO_2). Much of the world's current demand for thorium metal and its compounds is satisfied by mining placers along India's Malabar Coast, where wave action deposits monazite as a coarse yellow-to-brown

sand on beaches. Other ores of thorium, not commercially mined, are the oxide mineral thorianite (ThO_2) and the silicate mineral thorite ($ThSiO_4$).

Monazite beach sands are readily mined with conventional placer mining equipment and procedures. The dredged monazite is admixed with a variety of other minerals, including silica, magnetitie, ilmenite, zircon, and garnet. Concentration is accomplished by washing out lighter minerals in shaking tables and passing the resulting monazite fraction through a series of electromagnetic separators, which separate monazite from other minerals by virtue of their different magnetic permeabilities.

Although monazite is very stable chemically, it is susceptible to attack by both strong mineral acids (e.g., sulfuric acid, H_2SO_4) and alkalies (e.g., sodium hydroxide, NaOH). In the acid treatment, finely ground monazite sand is digested at 155° to 230° C (310° to 445° F) with highly concentrated (93 percent) H_2SO_4. This converts both the phosphate and the metal content of the monazite to water-soluble species. The resulting solution is contacted with aqueous ammonia, first precipitating hydrated thorium phosphate as a gelatinous mass and then metathesizing the thorium phosphate to thorium hydroxide. Finally, the crude thorium hydroxide is dissolved in nitric acid to produce a thorium nitrate-containing feed solution suitable for final purification by solvent extraction. In alkaline digestion, finely ground monazite sand is carefully treated with a concentrated NaOH solution at 138° C (280° F) to produce a solid hydroxide product. Any one of several mineral acids is then used to dissolve this solid residue. For example, treatment with hydrochloric acid yields a solution of thorium and rare-earth chlorides. Conventionally, thorium is partially separated from the rare earths by addition of NaOH to the acidic chloride solution. The crude thorium hydroxide precipitate is then dissolved in nitric acid for final purification by solvent extraction.

For the purification of thorium from residual rare earths and other contaminants present in nitric acid feed solutions, the crude thorium nitrate concentrate is usually contacted with a solution of tributyl phosphate diluted by a suitable hydrocarbon. The resulting organic extract, containing the thorium (and any uranium that may be present), is then contacted countercurrently with a small volume of nitric acid solution in order to remove contaminating rare earths and other metallic impurities to acceptable levels. Finally, the scrubbed tributyl phosphate solution is contacted with a dilute nitric acid solution; this removes, or strips, thorium from the organic solvent into the aqueous solution while retaining uranium (if present) in the organic phase. Thermal concentration of the purified thorium nitrate solution yields a product suitable for the fabrication of gas mantles (see below Chemical compounds). The nitrate can also be calcined to ThO_2, which is incorporated into ceramic fuel elements for nuclear reactors or is converted to thorium metal.

Powdered ThO_2 can be fluorinated with gaseous hydrogen fluoride (HF), yielding thorium tetrafluoride (ThF_4). The metal is obtained by the Spedding process, in which powdered ThF_4 is mixed with finely divided calcium (Ca) and a zinc halide (either zinc chloride or zinc fluoride) and placed in a sealed, refractory-lined "bomb." Upon heating to approximately 650° C (1,200° F), an exothermic reaction ensues that reduces the thorium and zinc to metal and produces a slag of calcium chloride or calcium fluoride:

After solidification, the zinc-thorium alloy product is heated above the boiling point of zinc (907° C, or 1,665° F) but below the melting temperature of thorium. This evaporates the zinc and leaves a highly purified thorium sponge, which is melted and cast into ingots.

When bombarded by thermalized neutrons (usually released by the fission of uranium-235 in a nuclear reactor), thorium-232 is converted to thorium-233. This isotope decays to

protactinium-233, which in turn decays to uranium-233. The fissile properties of uranium-233 can be utilized immediately or after recovery from the irradiated reactor fuel.

Uranium-233 can be recovered and purified from neutron-irradiated thorium reactor fuels through the thorium extraction, or Thorex, process, which employs tributyl phosphate extraction chemistry. Irradiated fuel, containing either thorium metal or oxide, is dissolved in nitric acid containing a small amount of fluoride ion. Uranium-233 and thorium are coextracted into a tributyl phosphate solution, which is then contacted with an aluminum nitrate solution to remove traces of accompanying fission products. Dilute nitric acid is used to preferentially remove thorium from the scrubbed organic phase. Uranium-233 remaining in the tributyl phosphate solvent is stripped into acidified water; the resulting strip solution is passed through an ion-exchange resin bed in order to concentrate and purify the uranium-233.

Thorium is reported to alloy readily with many elements, including aluminum, beryllium, bismuth, boron, cobalt, copper, gold, iron, lead, magnesium, mercury, molybdenum, nickel, platinum, selenium, silver, sodium, tantalum, tungsten, and zinc. Some thorium is alloyed with magnesium metal to produce a material of increased high-temperature strength.

Aqueous solutions of highly purified thorium nitrate, $Th(NO_3)_4$, are produced when thorium ores are processed (see above Extraction and refining). The nitrate is extensively used in the commercial production of gas mantles. Such mantles are made by impregnating cotton or synthetic fibers with a 25 to 50 percent solution of $Th(NO_3)_4$ containing 0.5 to 1 percent each of thorium sulfate and cerous nitrate. The impregnated fibres are treated with aqueous ammonia, producing thorium hydroxide, $Th(OH)_4$, and this compound is calcined to produce ThO_2. The latter substance, when heated, emits brilliant white light. The added cerous nitrate improves spectral emission properties, while the small amounts of thorium sulfate yield mantles with improved mechanical properties. The only other thorium compound of any industrial significance is ThO_2, known as thoria. For nuclear applications, thoria is prepared by calcination of thoroughly purified $Th(NO_3)_4$. Thoria also finds some application as a refractory material in various high-temperature processes.

Thorium resources, mining, milling, and fuel fabrication

Thorium is widely distributed in nature with an average concentration of 10 ppm in earth's crust in many phosphates, silicates, carbonates and oxide minerals. Natural thorium is present as nearly 100% 232Th isotope. In general, thorium occurs in association with uranium and rare earth elements in diverse rock types: as veins of thorite, thorianite, uranothorite and as monazite in granites, syenites, pegmatites and other acidic intrusions. Monazite is also present in quartz-pebble conglomerates sand stones and in fluviatile and beach placers. In addition, thorium is also found as an associate element with rare earth elements bearing bastnaesite in carbonatites.

The present knowledge of thorium resources in the world is limited and incomplete because of the relatively low-key exploration efforts arising out of insignificant demand. Apart from its main use in nuclear energy, as 'fertile' material, thorium finds limited application in nonnuclear areas, mainly as thorium nitrate for gas mantles and to a very limited extent as thorium oxide refractory, catalyst (for synthesis of either methane or mixtures of saturated and unsaturated hydrocarbons from mixtures of CO and H2), thoriated tungsten welding rods and in magnesium-based alloys.

The largest known reserves of thorium are contained in the beach and inland placer deposits

of monazite, a mixed phosphate mineral with chemical formula: (RE/Th/U) PO4. Monazite is a primary source of light REE and thorium and a secondary source of phosphate and uranium. The total known world reserves of thorium in the Reasonably Assured Reserves (RAR) and Estimated Additional Reserves (EAR) categories are in the range of 2.23 million tonnes and 2.13 million tonnes respectively as shown in Table A.5-1.

In the RAR category, the deposits in Brazil, Turkey and India are in the range of 0.60, 0.38 and 0.32 million tonnes respectively. The thorium deposits in India has recently been reported to be in the range 0.65 million tonnes.

Table A.5-1 – Estimated thorium reserves (tones of Th metal)		
Country	Reasonably Assured Reserves (RAR)	Estimated Additional Reserves (EAR)
Australia	19,000	-
Brazil	606,000	700,000
Canada	45,000	128,000
Greenland	54,000	32,000
Egypt	15,000	309,000
India	319,000	-
Norway	132,000	132,000
South Africa	18,000	-
Turkey	380,000	500,000
United States	137,000	295,000

In ^{232}Th–^{233}U fuel cycle, much lesser quantity of plutonium and long-lived Minor Actinides (MA: Np, Am and Cm) are formed as compared to the ^{238}U–^{239}Pu fuel cycle, thereby minimizing the radiotoxicity associated in spent fuel. However, in the back end of ^{232}Th–^{233}U fuel cycle, there are other radionuclides such as ^{231}Pa, ^{229}Th and ^{230}U, which may have long-term radiological impact. Thorium fuels have been made in the past, and for oxide, one presumably suitable technology would be similar to that already established industrially for uranium oxide and MOX fuels formed from pellets in tubular cladding. Separate plant would be needed to avoid cross contamination, and the optimum conditions could well be rather different, but no serious difficulties seem likely in the first cycle. If interest passes to nitride or other less familiar forms, then industrialization is likely to need a more radical development program.

Fabrication of high density sintered ThO_2 pellets for the ThO_2 bundles used for flux flattening of the initial Core of PHWRs and as blanket in FBTR, is carried out by the conventional Powder Metallurgy technique of cold compaction and high temperature sintering either in reducing or in oxidizing atmosphere as shown in Figure A.5-4.

No special problems are expected in manufacturing technology of mixed oxide thorium uranium (or plutonium) pelletelized fuel. ThO_2-based mixed oxide fuels are expected to have superior thermophysical properties, such as higher melting point, better thermal conductivity and lesser release of fission gas as compared to UO_2-based mixed oxide. For high burnup, the fission gas release, the fuel swelling, and the fuel centerline temperature are lower for mixed thorium-uranium oxide compared to the standard UO_2 pellets.

The coated fuel particles of thoria, mixed thoria urania, thorium di–carbide and mixed thorium uranium di–carbide have demonstrated excellent performance in the past in the high temperature gas cooled reactors (HTGR) in Germany, USA and UK.

Commercial nuclear fuel fabrication facilities in the United States must obtain licenses from the NRC to manufacture, produce, receive, acquire, own, possess, use, or transfer byproduct, source, and special nuclear material (10 CFR 30.3; 10 CFR 40.3; 10 CFR 70.3). Each license specifies the authorized byproduct, source, and special nuclear materials, their chemical and/or physical forms, and the maximum quantity of each material that the licensee is allowed to possess at any one time.

Figure A.5-4– *Flowsheet for ThO2 fuel bundle fabrication.*

Each license further designates the purposes for which the authorized materials may be used. Hence, if DOE elects to transfer depleted UF_6 to a commercial nuclear fuel fabrication facility for conversion to UO_2, the NRC license held by the facility would need to authorize receipt, storage, conversion, and transfer of depleted uranium.

Types of fuels and fuel elements

There is a big diversity of thorium-based nuclear fuels and fuel elements depending on the type of reactor. Except for the molten salt breeder reactor (MSBR), which uses mixed fluoride in liquid form as fuel and primary coolant, all other reactors use solid fuels in the form of tiny "ceramic fuel microspheres" (100–1000 µ), "ceramic fuel pellets" or "metallic alloy fuel rods".

Fuel fabrication

The radiotoxicity, quantity and form of the fuel material will determine the specific selection of process and the mode of fabrication to be employed for either first cycle or recycled fuel. Fuels containing naturally occurring 'fissile' ^{235}U in combination with 'fertile' ^{238}U or ^{232}Th, emitting only alpha particles of relatively low specific activity, can be manufactured by the so-called 'contact operations' where the operator has direct contact with the fuel material. However, process operations that involve generation and handling of fine powders of ^{235}U, ^{238}U or ^{232}Th bearing fuels are carried out in ventilated enclosures, such as glove boxes, for minimizing radioactive aerosol. Because both uranium and thorium are pyrophoric enclosures need to have atmosphere control for handling ^{235}U, ^{238}U or ^{232}Th bearing materials.

ThO_2, ThO_2–UO_2 and ThO_2–PuO_2 fuels have been manufactured in both 'particulate' (microspheres) and 'pellet' forms for use in water cooled reactors. ThO_2, UO_2 and PuO_2 are completely solid soluble and have very similar thermodynamic and thermophysical properties. The manufacturing processes of thoria based fuels are, therefore, similar to that of the well-established processes for fabrication of UO_2 and mixed oxide fuels.

However, as mentioned earlier, a special feature of ^{232}Th–^{233}U fuel cycle is the high gamma dose associated the daughter products of ^{232}U, which is always associated with ^{233}U and the high specific radioactivity of ^{233}U. Hence, handling of ^{233}U bearing materials, like ^{239}Pu and its higher isotopes, require additional safety controls.

The major fabrication campaigns reported, so far, for ThO_2–based water cooled reactor fuels are based on the conventional 'powder-pellet' route, involving cold pelletisation of fine powder or powder mixtures, followed by sintering. The 'powder pellet' route is suitable for fabrication of high-density fuel pellets but has the disadvantage of 'radiotoxic dust hazard' as it involves handling of fine fuel particles. Further, fine powders have poor flowability, which makes automation and remote fabrication somewhat difficult.

The following techniques have been developed so far for manufacturing ThO_2 and thoria based mixed oxide fuels:

(1) **'Powder-pellet' route**: for preparation of high density fuel pellets, using ThO_2, UO_2 and PuO_2 powders as starting materials; the fuel pellet stacks are encapsulated in cladding tubes.

(2) **'Vibro-sol' route**: for preparation of fuel microspheres using nitrate solutions of uranium, plutonium and thorium as starting materials and adapting 'ammonia external gelation' or 'ammonia internal gelation' process for obtaining hydrated gel microspheres; the microspheres are sintered and vibro packed in cladding tubes followed by encapsulation.

(3) **'Sol-gel microsphere pelletisation'**: using dust-free and free-flowing sol gel derived oxide fuel microspheres for direct pelletisation and sintering.

(4) **'Impregnation technique'**: where (a) partially sintered $ThO2$ pellets of relatively low density ($\leq 75\%$ theoretical density) or (b) 'porous' $ThO2$ microspheres are vacuum impregnated in uranyl nitrate ('U' as 233U) or Pu–nitrate solution followed by calcination and sintering to form high density $ThO2$-based mixed oxide fuel pellets, which are encapsulated in cladding tubes.